Argonne NATIONAL LABORATORY | NREL NATIONAL RENEWABLE ENERGY LABORATORY | OAK RIDGE National Laboratory

WPI | Michigan Technological University | UC San Diego

ReCell ADVANCED BATTERY RECYCLING

RECELL CENTER FOCUS AREAS:

-DESIGN FOR RECYCLE
-MODELING AND ANALYSIS

Project ID: bat435

J. SPANGENBERGER
Argonne National Laboratory

June 11, 2019

2019 DOE Vehicle Technologies Office
Annual Merit Review

U.S. DEPARTMENT OF ENERGY | Energy Efficiency & Renewable Energy
VEHICLE TECHNOLOGIES OFFICE

OVERVIEW

Timeline

- Project start: October 2018
- Project end: September 2021
- Percent complete: ~15%

Barriers

- Recycling and Sustainability
  - Cost to recycle is currently 5-15% of battery cost
  - Material shortage (Li, Co, and Ni)
  - Varying chemistries result in variable backend value

Budget

Year 1	$4,615k
Argonne	*$2650k*
NREL	*$965k*
ORNL	*$550k*
UCSD	*$150k*
WPI	*$150k*
MTU	*$150k*

Partners

- Argonne National Laboratory
- National Renewable Energy Laboratory
- Oak Ridge National Laboratory
- University of California, San Diego
- Worcester Polytechnic Institute
- Michigan Technological University

RELEVANCE - RECELL CENTER

Objective:

Foster the development of cost-effective and environmentally sound processes to recycle lithium-ion batteries

Bring together battery recycling experts to bridge technical and economic gaps to enable industry adoption

Impact:

Reduced cost of ownership and helping to drive battery costs to DOE's $80/kWh goal

Reduce primary material production to avoid material shortages and reliance upon foreign sources, increasing our nation's energy security

Minimize environmental impacts of the battery life cycle

MILESTONES

Q1 (Center) Establish the battery recycling center’s mission and include its targets and goals

✓ COMPLETED 12/21/18:

“Decrease the cost of recycling lithium ion batteries to ensure future supply of critical materials and decrease energy usage compared to raw material production”

Q2 (NREL) Provide an initial progress report on roll-to-roll relithiation

✓ COMPLETED 3/29/19: Roll-to-roll relithiation work is progressing and the concept is currently being tested using coin cells

Q3 (ORNL) Provide an initial progress report on design for recycle initiative

In progress

Q4 (Argonne) Establish the ReCell Center’s Battery Recycling Laboratory and Scale-up Facility

In progress

COLLABORATION AND ACKNOWLEDGEMENTS

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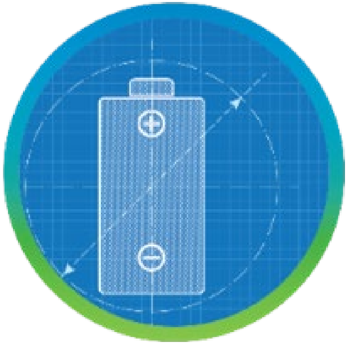
DESIGN FOR RECYCLING

FOCUS LEAD: JIANLIN LI

APPROACH – DESIGN FOR RECYCLING

Current batteries are designed with performance in mind, without much thought towards end-of-life. Designing new batteries with consideration of end-of-life can improve recyclability. But to keep the batteries marketable, the center will develop new designs that trade minimal loss in energy-density performance for the ability to use lower-cost, new recycling processes.

- Cell Design
  - Pouch cell, J. Li (ORNL)
  - Cylindrical cell, A. Jansen (Argonne)



CELL DESIGN

Fabricated NMC622 (3 mAh/cm^2) Cathode and Capacity-matched Graphite Anode

Anode: LN3174-62-4 & 63-1 (double-sided)
91.83 wt% Superior Graphite SLC1520P
2 wt% Timcal C-45
6 wt% Kureha 9300 PVDF Binder
0.17wt% Oxalic Acid
ReCell, 18650 cell build with cathode [LN3174-64-3&65-3]
Prod: SLC1520P, Lot#: Lot#: 022626-376-551
"SS" = single sided, "DS" = double sided -> ***CALENDERED***
Cu Foil Thickness: 10 µm
Total Electrode Thickness: 180 µm (DS)
SS Coating Thickness: 85 µm (SS)
Porosity: 32.2 %
Total SS Coating Loading: 12.49 mg/cm^2
Total SS Coating Density: 1.47 g/cm^3
Estimated SS Areal Capacity: 3.78 mAh/cm^2
[Based on rev. C/10 of 330 mAh/g for 0.005 to 1.5 V vs. Li]
Made by CAMP Facility

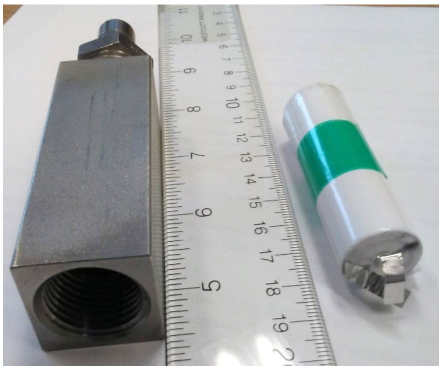
Cathode: LN3174-64-3 & 65-3 (double-sided)
90 wt% Targray NMC622
5 wt% Timcal C-45
5 wt% Solvay 5130 PVDF Binder
ReCell, 18650 cell build with anode [LN3174-62-4&63-1]
Prod: Targray SNMC03006, Lot#: TBD
"SS" = single sided, "DS" = double sided -> ***CALENDERED***
Al Foil Thickness: 20 µm
Total Electrode Thickness: 165 µm (DS)
SS Coating Thickness: 73 µm (SS)
Porosity: 31.2 %
Total SS Coating Loading: 20.43 mg/cm^2
Total SS Coating Density: 2.82 g/cm^3
Estimated SS Areal Capacity: 3.11(3.27) mAh/cm^2
[Based on rev. C/10 of 169(178) mAh/g for 3.0 to 4.2(4.3) V vs. Li]
Made by CAMP Facility

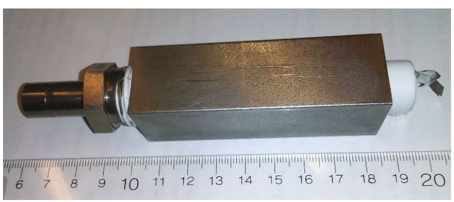
Electrodes fabricated at CAMP Facility (Argonne) and BMF (ORNL)

CELL DESIGN

Designed and Fabricated Cylindrical and Prismatic Cells

Cylindrical cell design at Argonne



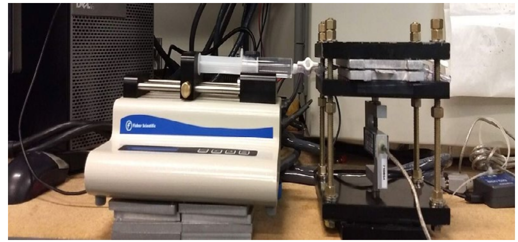


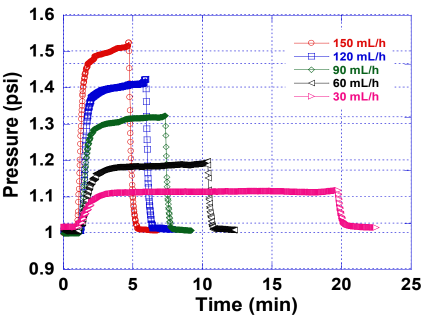
Prismatic cell design at ORNL

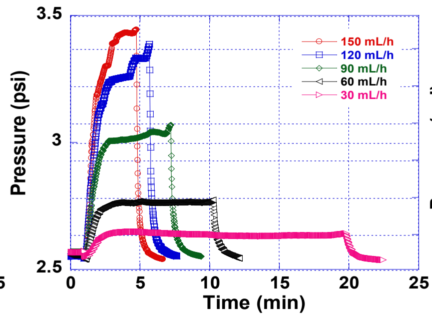
- Cylindrical and prismatic cells with ports were designed and fabricated for cell rejuvenation
- Significant efforts were paid to seal the assemblies and ports

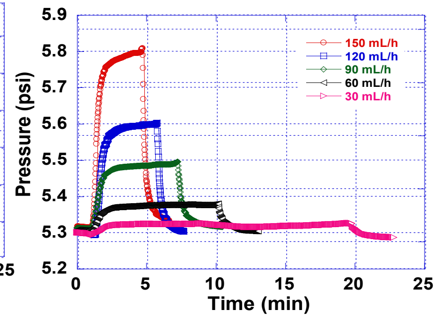
CELL DESIGN

Characterized Flow-Pressure Relation in Prismatic Cells









- A test fixture was built to measure cell pressure vs flow rate.
- Cell pressure increased with increasing flow rate and initial compression.
- High initial compression may force part of solvent to flow between the jelly roll and pouch material.

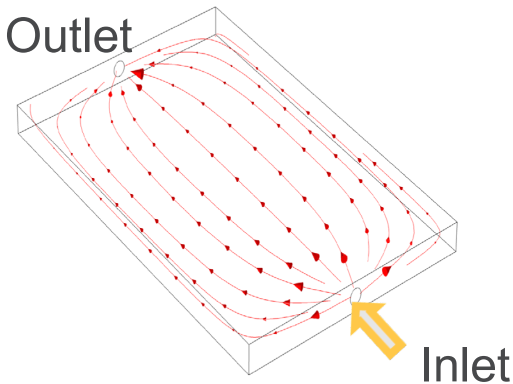
CELL DESIGN

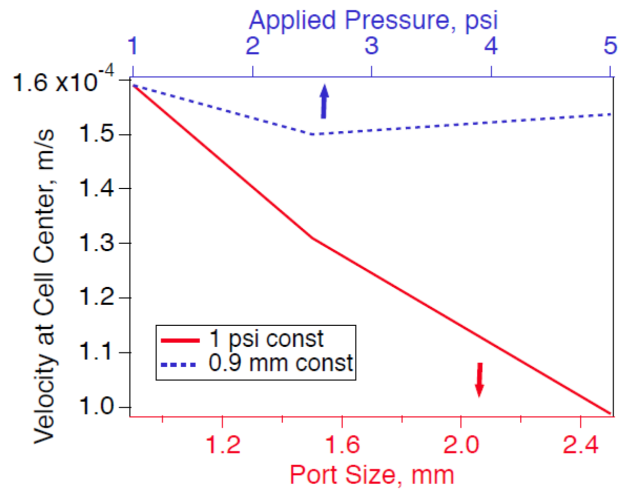
Velocity dependence on inlet size and applied pressure

- Ports are placed on the opposite edges of the cell
- Flow rate: 1ccm flow

- Cell dimension: 58 mmx 90 mm x5.5 mm
- Cell thickness: 5.5 mm
- Inlet radii: 0.9 mm, 1.5 mm, 2.5 mm

Velocity streamlines in the cell



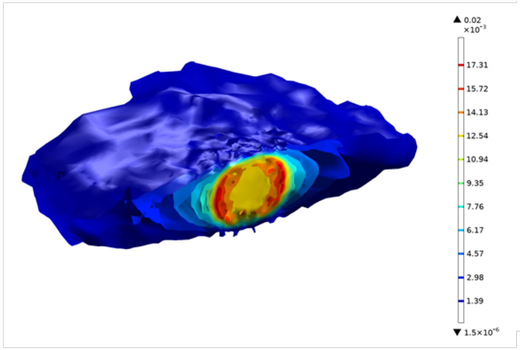


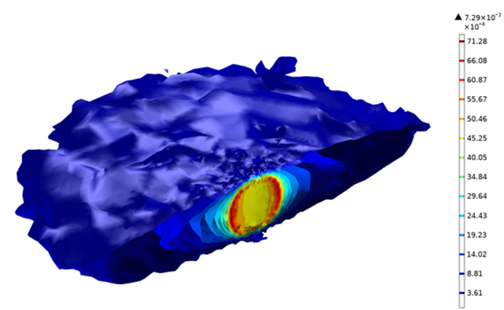
- Simplified model using average properties from cell components
- Extensive simulation required to incorporate individual layer properties

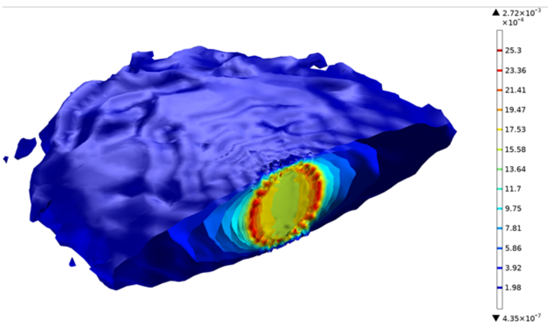
- Slower velocity at the center of the cell with large inlet size due to more liquid flows towards the sides
- Velocity independent of applied pressure

CELL DESIGN

Velocity distribution at the inlet (1 psi outer pressure)

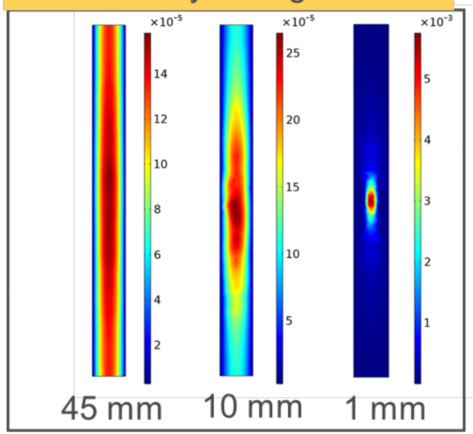






Outlet

Flow velocity through the cell



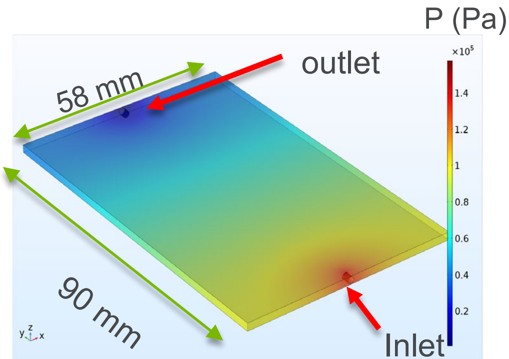
Inlet

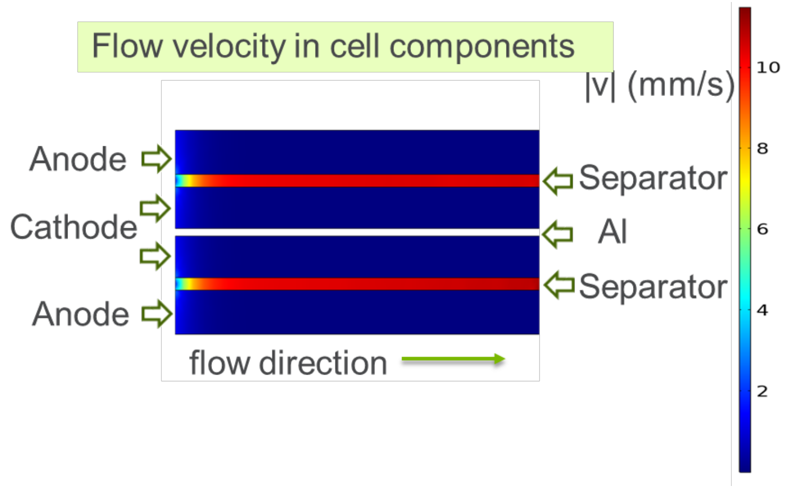
Radius: 0.9 mm

CELL DESIGN

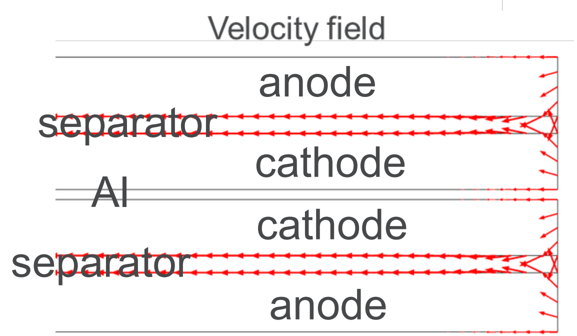
Simulate flow patterns and pressure gradient

- Flow rate:1 ccm
- Port size: Ø=0.9 mm
- No external compression





- Due to unavailable property of in-plane permeability, through-plane permeability of separator was used in simulation.
- Diethyl ether was used due to known viscosity as a function of pressure.
- Darcy's law $\kappa = \alpha \frac{\varepsilon^3 d^2}{(1-\varepsilon)^2}$
- High permeability separator provides "highways" for liquid flow.



REMAINING CHALLENGES AND BARRIERS

- Unavailable materials properties to accurately simulate flow patterns and cell pressure distribution
  - Permeability of liquids in cell components under compression
  - Viscosity of liquid under compression

- Solvent/electrolyte flow bypass of electrode assembly

- Non-uniform solvent/electrolyte flow distribution through cell assembly

FUTURE WORK

- Characterize flow-pressure in as-assembled cylindrical cells
- Characterize flow-pressure in cycled cylindrical and prismatic cells
- Estimate cell energy and power density in the new cell designs
- Screen solvents to effectively remove undesired compounds
- Rejuvenate harvested cycled electrodes and cycle in cells
- Evaluate cycle life benefit from electrolyte rejuvenation
- Assemble cylindrical and prismatic cells in proposed cell designs and cycle them

Any proposed future work is subject to change based on funding levels

SUMMARY

- Designed and fabricated cylindrical and pouch cells that allow for electrolyte rejuvenation
- Fabricated state of the art electrodes (NMC622 and graphite) for this work
- Began characterizing the flow-pressure relationship during removal of undesirable compounds and injecting fresh electrolyte
- Initiated evaluation of cell energy and power density in new cell designs

MODELING AND ANALYSIS FOR RECYCLING

FOCUS LEAD: QIANG DI

Argonne National Laboratory
NREL National Renewable Energy Laboratory
Oak Ridge National Laboratory
WPI
Michigan Technological University
UC San Diego
ReCell Advanced Battery Recycling
U.S. Department of Energy | Energy Efficiency & Renewable Energy
Vehicle Technologies Office

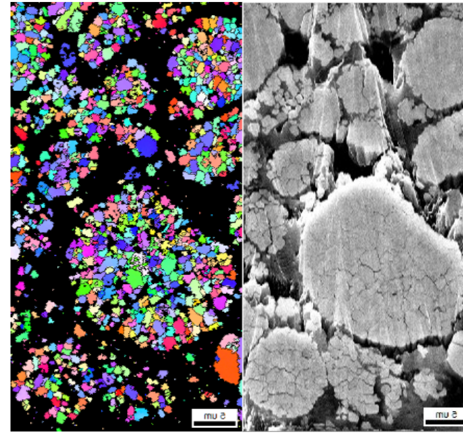
APPROACH – MODELING AND ANALYSIS

There are many potential recycling pathways for batteries that no longer meet their performance criteria. It would be expensive and time-consuming to explore all possible process options; researchers need tools to direct them to the most efficient and economic processes.

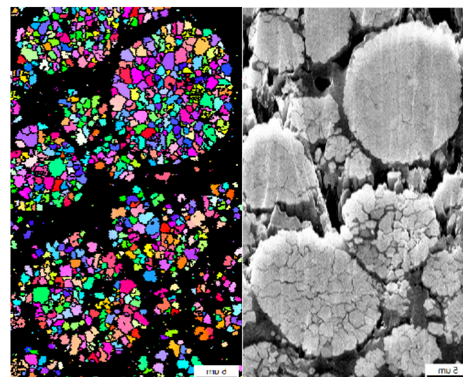
- Diagnostics on Aged Materials, S.Santhanagopalan (NREL)
- Thermal Analysis, M. Keyser (NREL)
- TEA/LCA Modeling (EverBatt), Q. Dai (Argonne)
- Supply Chain Analysis (LIBRA), M. Mann (NREL)

DIAGNOSTICS ON AGED MATERIALS

- Methodology to obtain grain orientation from cathode samples via electron back-scattering diffraction (EBSD) in place. Initial scanning transmission electron microscope (STEM) studies underway.
- Dry samples as well as first batch of cycled off-the-shelf $LiNi_{0.5}Mn_{0.3}Co_{0.2}O_2$ (NMC532) were characterized. Initial results for $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$ (NMC111) from Argonne being processed.
- EBSD patterns observed after 20% loss of capacity indicate no significant changes to particle size, grain orientation, etc. except for some hairline cracks. Some orientations show attrition; but nothing statistically significant yet.



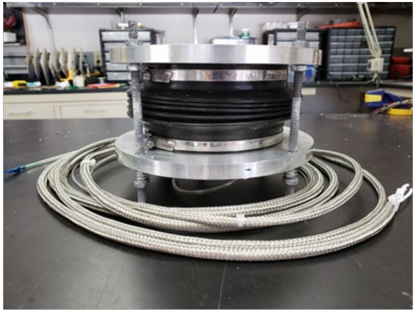
Uncycled NMC532



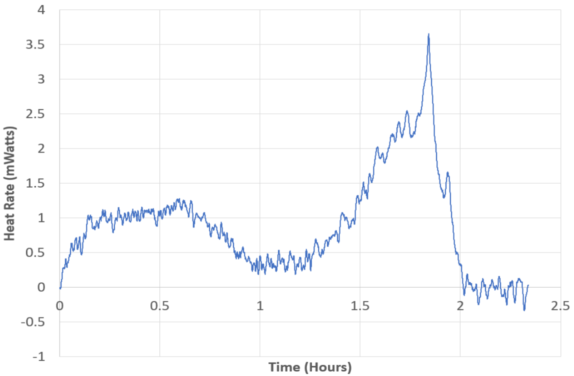
Cycled to 20% capacity loss

THERMAL ANALYSIS

- Thermally characterize existing lithium nickel manganese cobalt oxide (NMC) cathode compositions to understand how the thermal signature of a battery changes from the beginning of life to the end of life.
- Thermally characterize NMC/graphite cells with a known contaminant in both anode and cathode.
- Match the extent and type of thermal degradation to target recycling methods, based on load profiles, cycling windows, and cost metrics.



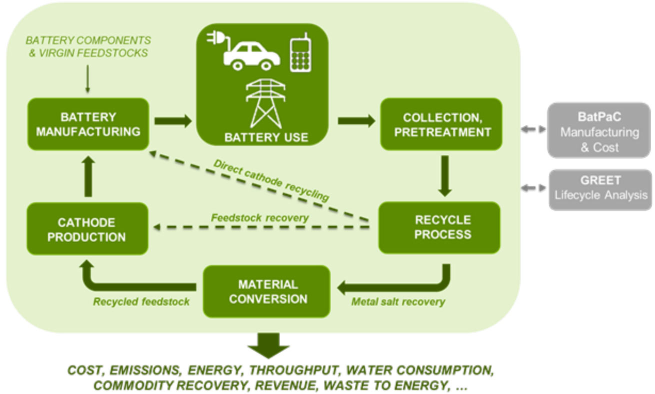
NREL's microcalorimeter.
Figure Credits, Josh Major, NREL



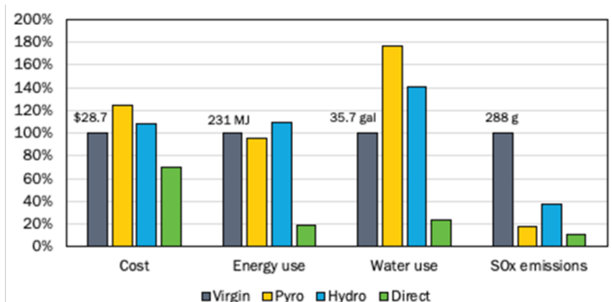
Heat generation of an NMC532/graphite cell under a C/2 constant current discharge.
Figure Credits: Matt Keyser, NREL

TEA/LCA MODELING (EVERBATT)

- Incorporated most recent BatPaC and GREET background data, and new industry information for recycling and cathode production processes.
- Model released in May 2019, available at: https://www.anl.gov/egs/everbatt
- Identified direct cathode recycling as a promising opportunity to reduce the cost and environmental burden of cathode materials.
- Expansion of model underway to enable modeling of new direct cathode recycling technologies, processes that recover anodes and electrolyte components, and design-for-recycle strategies.



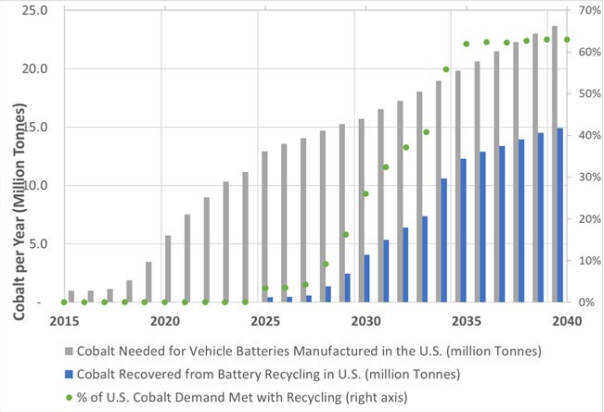
EverBatt Model Schematic

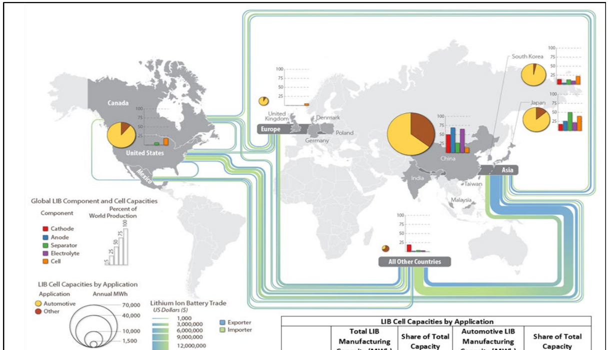


Cost and Environmental Impacts Comparison for 1kg Produced NMC111 Powder

SUPPLY CHAIN ANALYSIS (LIBRA)

- Updated 2014 analysis of supply chain of lithium-ion battery cells.
- Evaluated share of country-specific manufacturing for LIB for automotive, consumer electronic, and stationary energy storage markets.
- Determined fraction of the cobalt needed for vehicle manufacturing in the U.S. can be met with Li-ion battery recycling.
- Began development of Lithium Ion Battery Recycling Analysis (LIBRA) model – system dynamics model to study supply and demand of critical materials for batteries.





Note: Automotive manufacturing capacities were calculated based on the total production capacity of the studied facilities that make batteries for autos. Some of these facilities make batteries for autos and other applications.

LIB Cell Capacities by Application				
	Total LIB Manufacturing Capacity (MWh)	Share of Total Capacity	Automotive LIB Manufacturing Capacity (MWh)	Share of Total Capacity
China	118,234	62.3%	50,670	44.3%
Japan	22,479	11.8%	19,414	17.0%
S. Korea	18,547	9.8%	17,874	15.6%
U.S.	24,766	13.1%	22,016	19.2%
E.U.	2,626	1.4%	2,400	2.1%
Rest of World	3,110	1.6%	2,110	1.8%
Total	**189,762**	**100%**	**114,484**	**100%**

Sources of data: Trade Map 2017; BNEF 2017; Avicenne 2017; NREL analysis 2018

REMAINING CHALLENGES AND BARRIERS

- No correlations can be drawn between crystal structure and degradation mechanisms and recyclability.
- Typical contaminant levels may not be detectable during cycling in the thermal analysis effort.
- The availability of industrial data to populate the EverBatt model may be incomplete and/or difficult to obtain.
- The impact of policies and market dynamics on future battery material supply and demand may be hard to quantify.
- The assumptions that will be used to provide data based upon scaled-up ReCell process projects may be insufficient.

FUTURE WORK

- **Diagnostics on Aged Materials**: Build EBSD maps for cathode materials that are recovered/treated by different processes; Evaluate relationship between performance and grain size/orientation.
- **Thermal Analysis**: Measure the heat generation at the beginning and end of life and use the data as a baseline to assess the performance of future recycled material and processes; Use calorimetry to determine how contaminants on cathode and anode affect the performance of the cells.
- **TEA/LCA Modeling (EverBatt)**: Refine methodologies for techno-economic analysis and life cycle analysis; Continue to update model as new data/information become available; Continue to improve model usability.
- **Supply Chain Analysis (LIBRA)**: Evaluate investment decisions for recycling based on price of cobalt, policies, cost of recycling technology, and purity requirements; analyze material supply risk.
- **Focus Area**: Incorporate material characterization parameters into EverBatt.

Any proposed future work is subject to change based on funding levels

SUMMARY

- **Overarching goal**: Identify the optimal recycling processes for batteries, and inform and direct the R&D efforts under the ReCell Center.
- **Diagnostics on Aged Materials**: Identify chemical signatures of a battery corresponding to its efficiency losses; inform electrochemical relithiation conditions; match the extent and type of degradation to target recycling methods.
- **Thermal Analysis**: Understand how the thermal signature of a battery changes from the beginning of life to the end of life; thermally characterize cells with a known contaminant; match the extent and type of thermal degradation to target recycling methods .
- **TEA/LCA Modeling (EverBatt)**: Estimate the cost and environmental impacts of recycling processes and design-for-recycle strategies; identify hotspots and opportunities of improvement; identify possible barriers to process/design commercialization.
- **Supply Chain Analysis (LIBRA)**: Evaluate critical supply chain questions for battery recycling, including insights into material availability, the impact of demand from EV and stationary energy storage markets, and global economic competitiveness of battery manufacturing.

ReCell

ADVANCED
BATTERY RECYCLING

www.recellcenter.org

U.S. DEPARTMENT OF
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VEHICLE TECHNOLOGIES OFFICE

RESPONSE TO REVIEWERS

- **New Project FY19**